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A global emission inventory of carbonaceous aerosol from historic records of fossil fuel and biofuel consumption for the period 1860–1997

C. Junker and C. Liousse

Laboratoire d'Aérologie (URA CNRS 354), Université Paul Sabatier, Toulouse, France

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Correspondence to: C. Junker (junc@aero.obs-mip.fr)

Abstract

Country by country emission inventories for carbonaceous aerosol for the period 1860 to 1997 have been constructed on the basis of historic fuel production, use and trade data sets published by the United Nation's Statistical Division UNSTAT (1997), Etemad et al. (1991) and Mitchell (1992, 1993, 1995). The inventories use emission factors variable over time, which have been determined according to changes in technological development.

The results indicate that the industrialisation period since 1860 was accompanied by a steady increase in black carbon (BC) and organic carbon (OC) emissions up to 1910. The calculations show a moderate decrease of carbonaceous aerosol emissions between 1920 and 1930, followed by an increase up to 1990, the year when emissions began to decrease again.

Changes in BC and OC emissions prior to the year 1950 are essentially driven by the USA, Germany and the UK. The USSR, China and India become substantial contributors to carbonaceous aerosol emissions after 1950.

Emission maps have been generated with a $1^{\circ} \times 1^{\circ}$ resolution based on the relative population density in each country. They will provide a helpful tool for assessing the effect of carbonaceous aerosol emissions on observed climate changes of the past.

1 Introduction

BC and OC aerosol is increasingly recognised to have a major effect on regional and global radiative balance and climate (Jacobson et al., 2002).

The effects of anthropogenic particulate emissions in the atmosphere are well documented in the literature (Houghton et al., 2001; Seinfeld and Pandis 1998; Turco, 1997; Johnson et al., 2004), but the magnitude and even the sign of the effect are still uncertain. The abilities to absorb and scatter incident radiation are key properties which influence the magnitude and sign of their effects.

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While scattering aerosol generally contributes to tropospheric cooling, absorbing aerosol causes atmospheric warming through the direct aerosol effect, i.e. the transmission of absorbed heat into the atmosphere (Haywood and Ramaswamy, 1998; Jacobson, 2001), the semi-direct effect, i.e. the dissipation of clouds through their warming due to the heat generated by absorbing aerosol within the cloud (Hansen et al., 1997; Ackerman et al., 2000; Johnson et al., 2004), and by reducing the albedo of snow and ice surfaces (Hansen, 2004). Most of OC aerosols are scatterers of radiation whereas BC is the most important absorbing aerosol in the atmosphere.

The climate forcing effect of carbonaceous aerosol has been investigated by relating BC and OC emissions to observed climate changes of the past by Feichter et al. (2004), who use the BC emission inventory given by Cooke et al. (1999) and Liousse et al. (1996) for the 1990s and 1980s. Recently two authors have calculated BC emissions for periods extending further into the past. Novakov et al. (2003) calculates BC emissions as far back as 1875 using global CO_2 emission data and applying to it the BC/CO_2 ratio documented for the UK. While this work is the first attempt to calculate BC emissions for the late 19th century, it does not include OC and does not provide the spatial distribution required for global climate modelling. Furthermore, there is no evidence for the underlying assumption that the BC/CO_2 ratio in the UK is representative for the entire world, since it is variable and depends on the burnt fuels as well as on the technology used in the combustion processes.

Ito and Penner (2005) have conducted a study of historic carbonaceous aerosol emissions for the period 1870 to 2000 which includes emissions from open biomass burning. Like Novakov et al. (2003), they determine BC and OC emissions prior to 1950 by means of scaling CO_2 emission data. However, they use the earliest BC/CO_2 ratio found for each country instead of the BC/CO_2 ratio of the UK in each year of the calculation. In this study emission factors (EF) are kept constant over time with the exception of EF_{BC} for Diesel for the period 1974 to 1996.

The work presented here consists of a new emission inventory for BC and OC aerosol reaching back to the year 1860. It is a new approach compared to the previ-

ous studies because it directly calculates BC and OC emissions from fuel consumption data and appropriate emission factors for BC and OC varying over time. The BC and OC emissions per country and emissions per 1°×1° grid cell presented in this work exist for the years 1860, 1870, 1880, 1890, 1900, 1910, 1922, 1928, 1937, and for the years from 1946 to 1997.

2 Methods

This work uses two different methods for the inventories for the periods 1950 to 1997 and 1860 to 1949.

For the years 1950 to 1997 the method of Cooke et al. (1999) is used, which is based on the United Nations Energy Statistics Database (1997) (thereafter called the UNSTAT database). At its core are two simplifying assumptions which allow to reduce the vast data on fuel consumption in the UNSTAT database and the available data on EF for different fuel/usage/technology combinations.

1. Fuels can be categorised into two principal usage classes and associated EF: a) industrial use with EF_{Ind} and b) domestic use with EF_{Dom}. Where the usage of the fuel is not known, a third class c) "combined use" can be defined with an EF as the geometric mean of EF_{Ind} and EF_{Dom}. This class can also be associated with a particular fuel usage (and corresponding EF) where such a usage is known as in the case of liquid fuel use by traffic.

2. The difference in EF occurring in identical fuel/usage combinations as a consequence of different technologies employed can be categorised into three classes: EF for a) industrialised countries, b) semi-developed countries, and c) developing countries.

A simplified description scheme of the method of Cooke et al. (1999) is shown in Fig. 1a. This method is in contrast to the one used by Bond et al. (2004), who use

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specific EF for each fuel/usage combination, taking account of the technology used in each particular case. This approach was not used in this work because the information it necessitates is not available for all countries in the present time and even less available for past periods since 1950.

For the years 1860 to 1949 the method of Cooke et al. (1999) was further simplified because data on fuel usage is not available for this period. Instead, fuel usage in terms of the three classes industrial, domestic and combined is estimated globally, guided by the extrapolation of trends evident from the UNSTAT database and by knowledge of past fuel usage in some particular cases. The global estimates of fuel usage are incorporated into the EF of each fuel for each year for which the inventory is calculated. A description scheme for this method is shown in Fig. 1b.

Another modification of the methodology of Cooke et al. (1999) for the years prior to 1950 follows from the fact that the differentiation between fuel use in industrialised, semi-developed and developing countries cannot be applied in the same way to the periods before and after World War II. Prior to World War II the industrial activity in today's developing countries was dominated by colonial powers who can be expected to have used similar technologies as in their homeland. While there may have been a difference between the industrial installations in colonies and those in the homeland, the difference in resulting emissions is assumed to be smaller than today's.

Furthermore, there is no justification for a threefold division of stages of development. Therefore, only two classes of development are considered before 1939, which are semi-developed and developing.

3 Determination of consumed fuels

3.1 The period 1950 to 1997

The UNSTAT database, used for the period 1950 to 1997, gives fuel production, import and export data for 185 countries, provided by national statistical offices. The database

also gives information about the sector of usage of the fuels in each country allowing the distinction between domestic, industrial or traffic usage. The sector of usage information stems from surveys conducted by different bodies or agencies in the various countries. However, for the period 1950 to 1969, the data base information about the fuel usage sector is incomplete. In cases where sector of usage information is missing, the average sector "combined" described in Sect. 2 is used for the calculations.

From 1970 onwards the database contains both fossil fuels and biofuels, whereas biofuels are not included for the period 1950 to 1969. The approximate biofuel consumption for each country prior to 1970 was calculated from the value given by UNSTAT for the year 1970 and a proportionality factor derived from the decrease of the world population towards earlier years as shown in Fig. 2.

3.2 The period 1860 to 1949

Fossil fuel use during the period 1860 to 1949 is inferred from annual fuel production data given by Etemad et al. (1991) for coal, crude oil and peat. Figure 3 shows good agreement between total global fuel consumption data given by UNSTAT and fuel production data by Etemad et al. (1991) for the period of overlap from 1950 to 1985.

The fuel consumption is determined by adjusting the production data listed by Etemad et al. (1991) for 138 countries by fuel import and export data of Mitchell (1992, 1993, 1995) for 60 countries. Where no import or export data is provided by Mitchell (1992, 1993, 1995), the production data given by Etemad et al. (1991) is taken as a proxy for fuel consumption. Biofuel consumption is determined as described in Sect. 3.1.

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4 Emission factors (EF)

4.1 Fossil fuel EF for the reference year

The reference year for the inventory calculations is 1997. The EF values chosen for this year are based on those given by Cooke et al. (1999). However, in three cases the EF given by Cooke et al. (1999) are modified.

- EF diesel: the value of 2 g/kg given for traffic use in industrialised counties is reduced to 1 g/kg according to Liousse et al. (2004). Moreover, EF for domestic use is decreased, whereas EF for industrial use is increased according to Guillaume and Liousse (2006)[1].
- EF refinery oil: the same EF as for Diesel are applied instead of 0.025 g/kg for all the 9 categories (Guillaume and Liousse, 2006[1]).
- EF lignite: the same EF as for hard coal are applied instead of the higher values given by Cooke et al. (1999). The studies by Bond et al. (2004) and EPA (1996) show a large spread in the EF of lignite, suggesting that there is not sufficient evidence to warrant for $EF_{lignite}$ being significantly different from $EF_{hard\ coal}$.

These adjustments are in broad agreement with the values given by Bond et al. (2004), who have published central values for total particulate matter (TPM) together with BC/TPM ratios from uncited literature. Their values are shown in Table 1a in comparison to the EF for diesel and coal for 1997 used in this work. Table 1b shows the EF chosen by Ito and Penner (2005), which are based on the work of Bond et al. (2004) and Streets et al. (2001).

[1] Guillaume, B. and Liousse, C.: Development of carbonaceous aerosol emission inventories from fossil fuel over Europe at continental scale with focus on traffic at national and regional scales, in preparation, 2006.

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Apart from the EF for diesel fuel, in this work preference is given to EF values given by Cooke et al. (1999) over those given by Bond et al. (2004) for two reasons. 1) Fuel usage (sectorisation) is considered in all of the EF values given by Cooke et al. (1999) whereas Bond et al. (2004) consider fuel usage mostly by applying a technology factor in the emission calculations; 2) Cooke et al. (1999) give references for the BC/TPM ratios chosen for the derivation of EF_{BC} from EF_{TPM}.

The EF are adjusted according to three fuel usage classes and three levels of development of the respective countries as described in Sect. 2. The group of developed countries is composed of the members of the Organisation for Economic Cooperative Development (OECD) in 1984. The distinction between semi-developed and developing countries is based on the gross national product per capita, which improves the classification by Cooke et al. (1999). The adjustment of the EF for usage and level of development resulted for each fuel type in a matrix of 9 EF, as shown in Table 1a for the examples hard coal and diesel.

4.2 Biofuel EF for the reference year

The emission factors chosen for biofuels are guided by a) a literature search (Table 2) with a preference of experimentally determined values over literature reviews, and b) from new measurements of emissions of different representative biofuels in a combustion chamber (H. Cachier, personal communication). The range of both the experimentally determined $EF_{biofuel}$ and the $EF_{biofuel}$ found in the literature extend over more than one order of magnitude. The variations of EF are due to the effect of devices used (different stove types/open fires), burning conditions and fuel moisture.

An appropriate mean value for the biofuel EF to be used in the emission calculations appears to be 0.75 g/kg_{dm}, which is in agreement with the literature review by Andreae and Merlet (2001). This value is used for domestic usage in developed countries. The same value is used for semi-developed countries, but it is increased to 0.90 g/kg_{dm} for developing countries, where worse conditions of burning can be expected.

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4.3 Changes of EF in the past

The EF need to be adjusted in order to reflect the different stages of technological development prior to 1997. Thus, individual sets of EF have been developed in this work for each year for which emissions are calculated. The adjustment of the EF according to changes in technological development and emission control is performed as follows: The industrial EF for coal in developed countries prior to 1997 is based on the EF of coal fired power plants. It is scaled to their thermal efficiency given by Etemad et al. (1991) for the years 1890 through 1980. The thermal efficiency is related to the emission factor because it is dependent (among other factors) on the completeness of the combustion. A less complete combustion will engender higher CO and particulate carbon emissions. This approach has been used for the first time by Pertuisot (1993) who scaled EF_{BC} starting from a base value of 0.24 g_C/kg_{dm} in 1980 given by W. Cooke (personal communication). The EF calculated by Pertuisot (1993) and the corresponding $EF_{industrial}$ used in this work are shown in Fig. 4 together with the $EF_{industrial}$ for hard coal given by Cooke et al. (1999) for the 1990 decade. A similar scaling is applied to the $EF_{industrial}$ for coal in developing countries, starting from the value given for developing countries by Cooke et al. (1999) attributed to the year 1997. However, the scaling factor is reduced in such a way that the $EF_{industrial}$ calculated for the year 1860 is similar albeit slightly higher (of the order 5% to 10%) in developing countries than in industrialised countries. This assumption seems reasonable since the technologies used at this time can be considered similar throughout the world, whereby in colonies (i.e. in today's developing countries) not the latest and most advanced technologies may have been employed.

The changes of the $EF_{traffic}$ for diesel oil in developed countries prior to 1997 are based on a study by Yanowitz et al. (2000) who report a 6.8 fold decrease of BC emissions at low altitude between the years 1974 and 1997 due to emission control measures. The $EF_{traffic}$ for diesel in developing countries is found in accordance with the method employed for coal by adjusting the scaling factor such that the EF prior to

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1974 in developing countries is within the range of 5% to 10% above the $EF_{traffic}$ in developed countries. The same scaling was applied to $EF_{traffic}$ for gasoline. The time series of the EF for coal and diesel is shown in Fig. 4 for industrialised and developing countries.

Domestic EF are deemed constant over time, except for the adjustment following from the assumption that today's industrialised countries can be considered to be semi-developed, and today's semi-developed countries to be developing countries prior to 1939.

4.4 Adjustment of the EF prior to 1950 according to the global estimate of fuel usage

Following the methodology described in Sect. 2, the effect of fuel use changes prior to 1959 on BC and OC emissions are taken into account as part of the global EF determination. This is done by calculating weighted averages of the domestic and the industrial EF of coal, peat and biofuel. The estimated proportion of domestic to industrial fuel usage in the past, guided by the back-extrapolation of UNSTAT data and inferences from literature about historic fuel use is shown in Fig. 5a.

The EF of crude oil from traffic usage is distinctly different from both industrial and domestic usage, thus requiring a three-fold sectorisation. The estimation of the usage sectors of crude oil, shown in Fig. 5b is based on an extrapolation from the sectorisation in the UNSTAT database constrained by the fact that 1) prior to the year 1900 there was no domestic heating with oil (the first central heating with oil in a private home was installed in 1920), and 2) that petrol usage by traffic was zero prior to 1885, the year in which the automobile was invented.

The particulate $EF_{traffic}$ of gasoline and of diesel differ by about one order of magnitude (Fig. 4). Hence the EF of crude oil is also dependent on the proportion to diesel to gasoline usage. The apportionment between gasoline and diesel consumption within traffic usage of crude oil was based on data from the U.S. Federal Highway Administration, 1995 (EPA/600/8-90/057F, 2002), shown in Fig. 6. The usage of US data for our global inventory may lead to a slight overestimation of emissions for the time prior

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to 1950. This is because the dieselisation of railway traffic and heavy road traffic may not have occurred as fast in other countries as in the U.S.

The EF for coal and crude oil used for the period 1860–1949, which incorporate both technical development and changes in fuel use over time, are shown in Fig. 7 for developed, semideveloped and developing countries. Note that the figure does not extend to years after 1960 since EF adapted for the UNSTAT database are used for the period 1950–1997 (Fig. 4).

5 Results

A combined BC and OC emission time series was constructed based on Etemad et al. (1991) and Mitchell (1992, 1993, 1995) for the period 1860 to 1937 and on UNSTAT for 1950 to 1997. No fuel production or consumption data is available for the time of World War II. Emissions have been calculated for the years 1946, 1947, 1948 and 1949 based on Etemad et al. (1991) and Mitchell (1992, 1993, 1995) and are available upon request to the authors. The global emissions of BC and OC aerosol are given in Table 3 for selected years and shown in Fig. 8 (BC only) for the period 1860 to 1997. The period of World War II is represented by a dashed line in order to highlight the uncertainty of the values. It is likely that BC emissions during this time were higher than suggested by the simple interpolation in Figs. 8 and 9, resulting from wartime activities such as combat and military transport.

The combined time series from 1860 to 1997 shows maxima in 1920 and 1990. The global emission decrease after 1920 appears to be mainly caused by decreasing emissions in the USA, the UK and Germany, shown in Figs. 9a and b. There are two probable reasons for these decreases: 1) the economic depression of the 1920s, and 2) technological developments, which lead to a decrease of the emission factors entered into the inventory calculations.

Figure 9c shows a rapid increase of BC emissions in India and China starting in the 1960s, whereas England and France show a levelling off or a decrease of BC emissions

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(Fig. 9b). Emissions in Germany do not level off but show a steep increase between the years 1950 and 1970. The proportion of emissions attributable to East Germany during this period is 62%.

Emission maps are drawn from BC and OC emissions in each country. The emission distribution within each country is calculated proportional to the population density. The population density map of 1984 of the Goddard Institute for Space Studies at Columbia University in New York GISS was used. Adjustments were made to reflect international border changes following the Second World War and the break-up of the Soviet Union. Figure 10 shows the spatial distribution of BC and OC emissions for the year 1997.

The emission map for BC (Fig. 10a) shows somewhat larger emission densities in Europe compared to Africa, whereas the difference between Europe and Africa is smaller on the emission map for OC. This is consistent with the facts that the proportion of biofuel to fossil fuel consumption is higher in Africa and that biofuel has a higher OC/BC emission ratio than fossil fuel.

The year 1985 shows globally a similar emission density of BC for compared to 1997, but a higher emission density in central Europe (map not shown). This may be explained by the low standard emission controls during the communist era.

Figures 11a and b show the BC emission density for the years 1970 and 1950. The figures indicate that from 1950 to 1970 the BC emission density increased mainly in Eastern Europe, and in semi-developed countries, e.g. India and China. Very small or no increases are seen for developing countries in South America and Africa.

The emission density map for 1910, shown in Fig. 11c, suggests that the BC emissions of industrialised countries in 1910 were similar to their emissions in 1950. This finding is consistent with the BC emissions per country shown in Figs. 9a (USA) and 9c (Germany, UK and France). The total global BC emissions for 1950 and 1910 are 3.63 Mt and 2.36 Mt respectively.

The main difference between emission densities in 1910 and 1860 (Fig. 11d) occurs in industrialised countries, since in developing countries of 1910 and 1860 emissions are nearly exclusively domestic and thus less variable over time.

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6 Conclusion

This study gives for the first time to our knowledge a spatialised inventory of BC and OC emissions for a period extending back to the beginnings of industrialisation based on historical fuel production data, and taking account of emission factor changes over time. For the period from 1980 to 2000, the combined global BC emissions from fossil fuel and biofuel use are similar to those calculated exclusively for fossil fuel by Novakov et al. (2003) (Fig. 8). Our fossil fuel emission estimates are thus lower than those of Novakov et al. (2003) for this period, but still higher than those given by Ito and Penner (2005). For the years 1950 to 1970 the emissions calculated here are higher than the estimates of both Novakov et al. (2003) and Ito and Penner (2005). However, our inventory is in approximate agreement with the one of Novakov et al. (2003) for the period 1875 to 1920. A possible explanation for the agreement before 1920 and disagreement afterwards could be given as follows. The simplifying assumption of Novakov et al. (2003) that the global BC/CO_2 emission ratio is similar to the one in the UK may be only applicable for the period before 1920, as BC emissions in the early industrial age were dominated by a small number of countries with similar industries.

The inventory presented here will enable to draw conclusions about the effect of particulate emissions on observed climate changes of the past and to complement studies of past climate forcing by means of ice cores, such as the studies performed by Rowntree (1998) and Legrand and De Angelis (1992).

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Table 1. (a) EF matrix for hard coal and diesel fuel and EF values given by Bond et al. (2004).

fuel	fuel usage (this work)				Bond et al. (2004)	
type	level of development	industrial	domestic	other/traffic	fuel usage	mean EF
	developed	0.15	1.39	0.46	power	0.006
hard coal	semi-developed	0.30	2.28	0.82	industry	0.610
	developing	1.1	2.28	1.58	traditional kiln	5.0
	industrialised	0.6	0.07	1	standards in place	0.99
diesel	semi-developed	0.87	0.09	2	standards beginning	2.31
	developing	2	0.35	5	superemitter	7.92

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Table 1. (b) EF for hard coal and diesel fuel used by Ito and Penner (2005).

fuel type	level of development	iron and steel industry	mineral industry	rest industries	residential	transport
hard coal	developed	0.3216	0.0046	0.0046	1.1250	0.4950
	semi-developed	0.6256	0.0729	0.0117	1.6068	0.4950
	developing	1.2172	1.1468	0.0293	2.2950	0.4950
					transport	
		agriculture		other sectors	road	rail
diesel	developed	2.4974		0.0653	1.1494	1.5325
	semi-developed	2.7245		0.4714	2.4691	1.5325
	developing	3.1786		3.4056	2.9515	1.5325

Table 2. Overview of the used literature for biofuel emission factors. In the studies where TPM was measured instead of BC or OC a factors of BC/TPM=12% and OC/TPM=40% was used, following Liousse et al. (1996).

	BC/dm [g/kg]	OC/dm [g/kg]	Reference		BC/dm [g/kg]	OC/dm [g/kg]	Reference
wood	0.59	1.96	Butcher and Sorenson (1979)	softwood	1.30	2.80	Muhlbaier and Williams (1982)
fuels	0.41	1.38	Butcher et Ellenbecker (1982)		2.48	8.26	Dasch (1982)
general	1.40	4.20	Turn et al. (1997)		0.72	2.40	Dasch (1982)
	1.70	4.30	Turn et al. (1997)		2.52	8.41	Radke et al. (1991)
	1.03	**2.96**	**average**		2.04	6.79	Radke et al. (1991)
					2.60	8.67	Turn et al. (1997)
residential	0.40	3.53	Brocard (1996)		1.25	4.17	Turn et al. (1997)
heating	0.70	4.20	Cooper (1980)		1.56	5.20	Butcher and Sorenson (1979)
and	1.20	4.20	Cooper (1980)		1.74	5.80	Patterson and MacMahon (1984)
cooking	0.04	0.12	Piispanen et al. (1984)		3.24	10.81	Patterson and MacMahon (1984)
	1.09	3.64	Butcher et al. (1984)		0.27	0.90	Butcher et Ellenbecker (1982)
	1.20	4.00	Dasch (1982)		1.23	4.10	Radke et al. (1991)
	1.48	4.92	Butcher et al. (1984)		4.21	14.04	Radke et al. (1991)
	0.92	3.08	Butcher et al. (1984)		0.32	2.30	EPA/600/R-00/052 (2000)
	0.62	2.08	Butcher et al. (1984)		0.24	1.63	EPA/600/R-00/052 (2000)
	0.60	2.00	Smith et al. (1983)		0.25	1.59	EPA/600/R-00/052 (2000)
	0.33	1.11	Zhang et al. (2000)		**1.62**	**5.49**	**average**
	1.32	4.40	Butcher and Sorenson (1979)				
	1.20	4.00	Smith et al. (1993); Butcher et al. (1984)	Charcoal	0.29	1.76	EPA/600/R-00/052 (2000)
	0.85	**3.18**	**average**	burning	0.20	1.80	Brocard (1996)
					0.96	3.20	Butcher et al. (1984)
hardwood	0.39	4.70	Muhlbaier and Williams (1982)		1.50	5.00	Smith et al. (1993); Butcher et al. (1984)
	0.57	1.89	Dasch (1982)		**0.74**	**2.94**	**average**
	0.45	1.50	Butcher and Ellenbecker (1982)				
	0.96	3.20	Butcher and Sorenson (1979)	Charcoal	0.34	3.22	Cachier (1996)
	0.59	**2.82**	**average**	making	0.07	0.23	EPA/600/R-99/109 (1999)
					0.96	2.80	Pennise et al. (2001)
					0.46	**2.09**	**average**

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Table 3. Total global BC and OC aerosol emissions from 1860 through 1997 on the basis of fossil fuel calculated from data of Etemad et al. (1991) for the period 1860–1937 and from UNSTAT for the period 1950–1997.

year	BC [Mt]	OC [Mt]	BC from biofuel [%]	OC from biofuel [%]
1860	0.72	1.76	49	61
1880	1.29	2.59	31	47
1900	2.39	4.04	19	34
1922	2.38	4.22	22	37
1937	2.02	3.95	28	43
1950	3.29	5.54	21	37
1970	4.99	8.03	22	42
1985	5.93	9.46	26	48
1997	5.77	9.57	31	56

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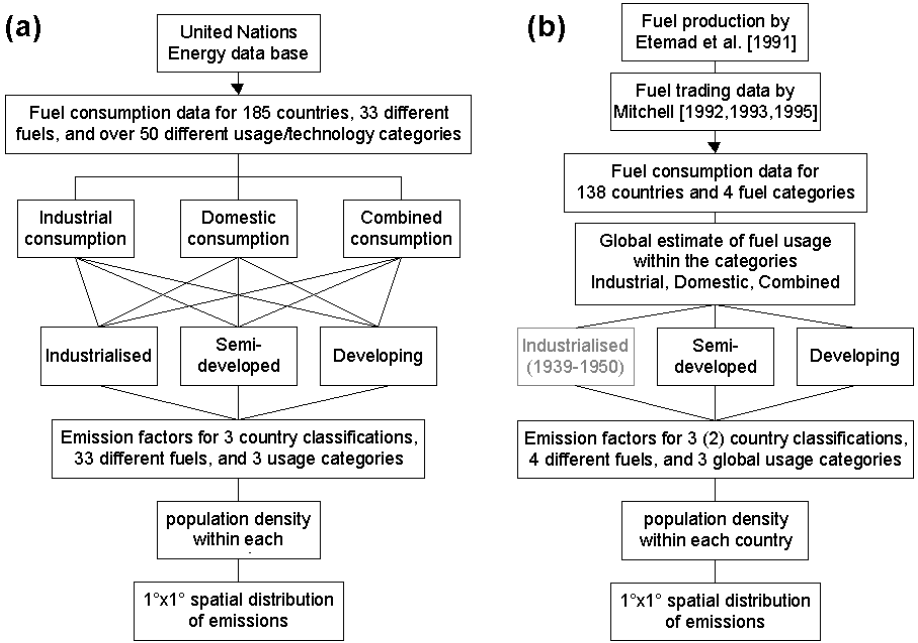

Fig. 1. Tree diagrams of the methods used for the inventory of **(a)** the period 1860–1949 and **(b)** the period 1950–1997.

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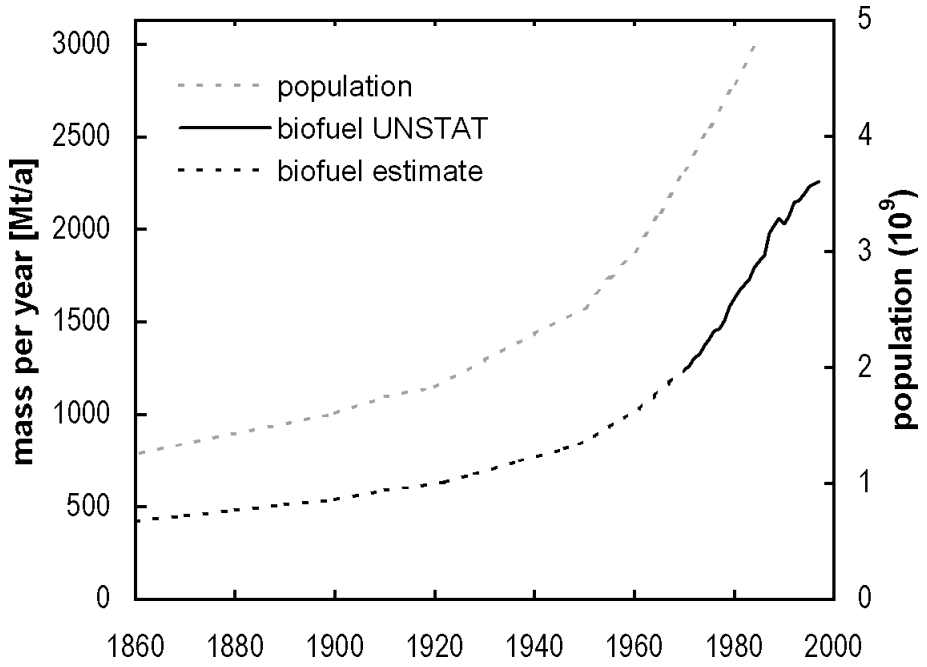

Fig. 2. Biofuel consumption from UNSTAT for the period 1970 to 1997 and biofuel consumption estimate according to world population growth.

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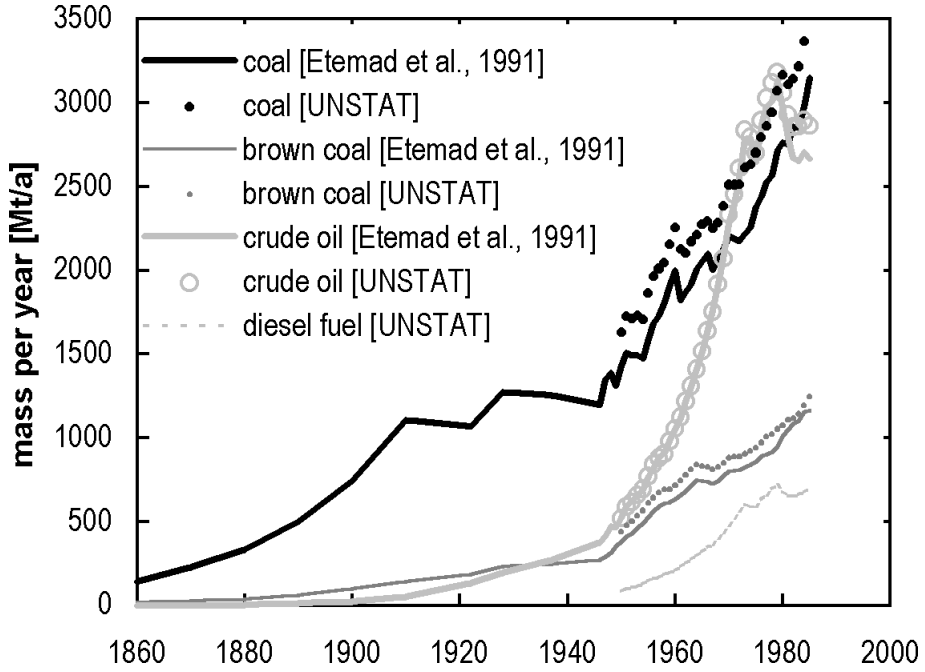

Fig. 3. Fuel production from Etemad et al. (1991) and fuel consumption of the United Nations Statistical division UNSTAT (1997).

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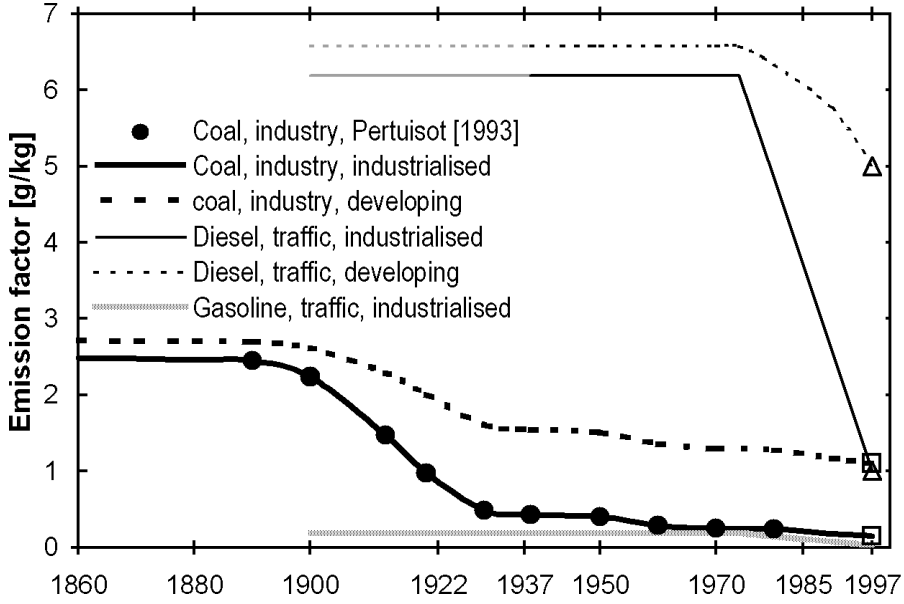

Fig. 4. BC emission factors for coal and liquid fuels used in this work and those given by Pertuisot (1993). The EF for the year 1997 (Cooke et al., 1999) are shown as square symbols (hard coal) and triangular symbols (diesel). The lines representing $EF_{traffic}$ for diesel oil are grey-shaded prior to 1938 because traffic using diesel engines was only experimental at this time. The years marked on the abscissa are years for which global emissions are presented in Table 3.

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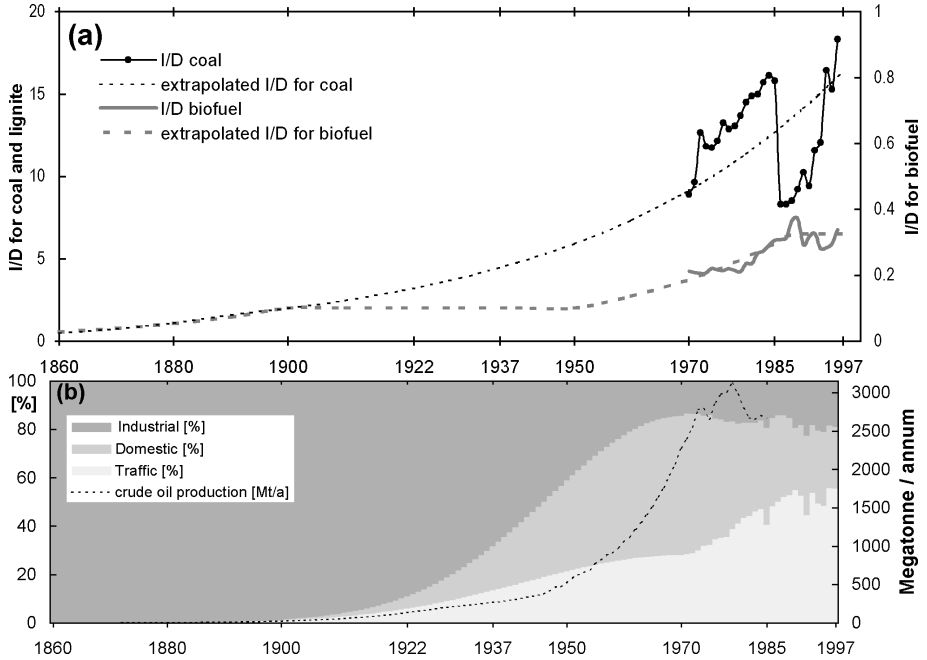

Fig. 5. (a) Ratio of Industrial use to Domestic use (I/D) of solid fuels from UNSTAT data 1970 to 1997 and extrapolation and estimation for the period 1860 to 1970. **(b)** Percentages of industrial, domestic and traffic use of crude oil from UNSTAT data for the period 1970 to 1997 and extrapolation and estimation for the period 1860 to 1970. The crude oil production, shown as a dashed line, is taken from Etemad et al. (1991). The years marked on the abscissa are years for which global emissions are presented in Table 3.

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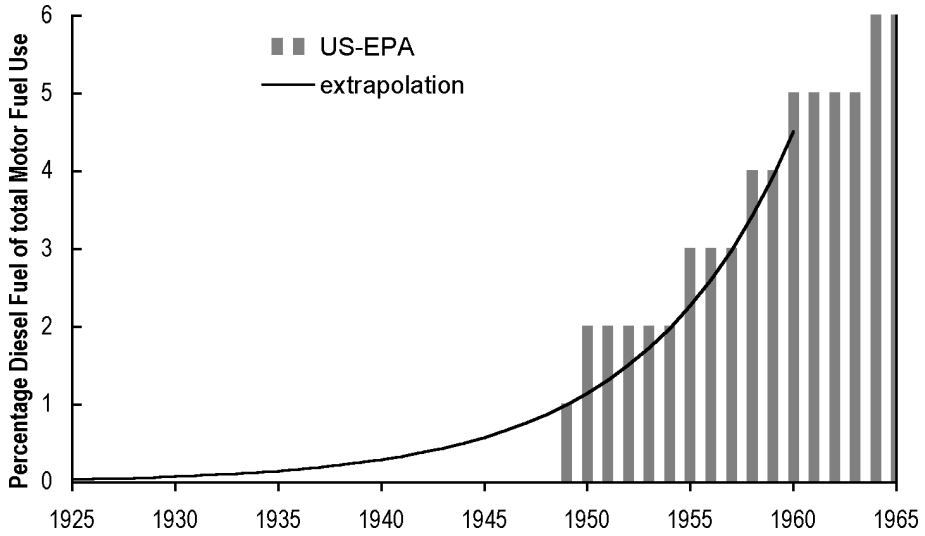

Fig. 6. Diesel fuel use relative to total motor fuel use given by EPA/600/8-90/057F (2002) and backward extrapolation used in this work.

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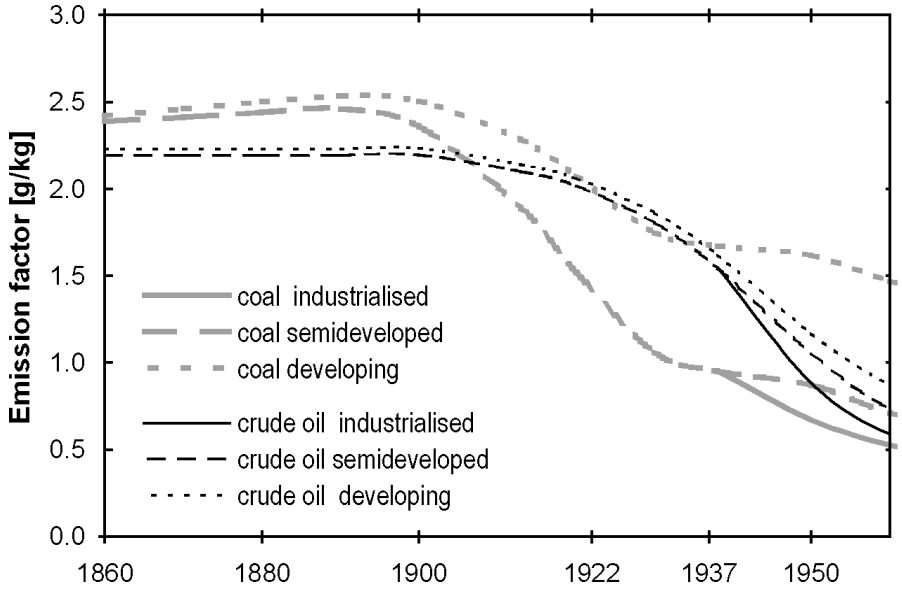

Fig. 7. BC emission factors for coal and crude oil according to changes in fuel usage over time, used for the period 1860 to 1949 with fuel consumption data based on Etemad et al. (1991) and Mitchell (1992, 1993, 1995). The years marked on the abscissa are years for which global emissions are presented in Table 3.

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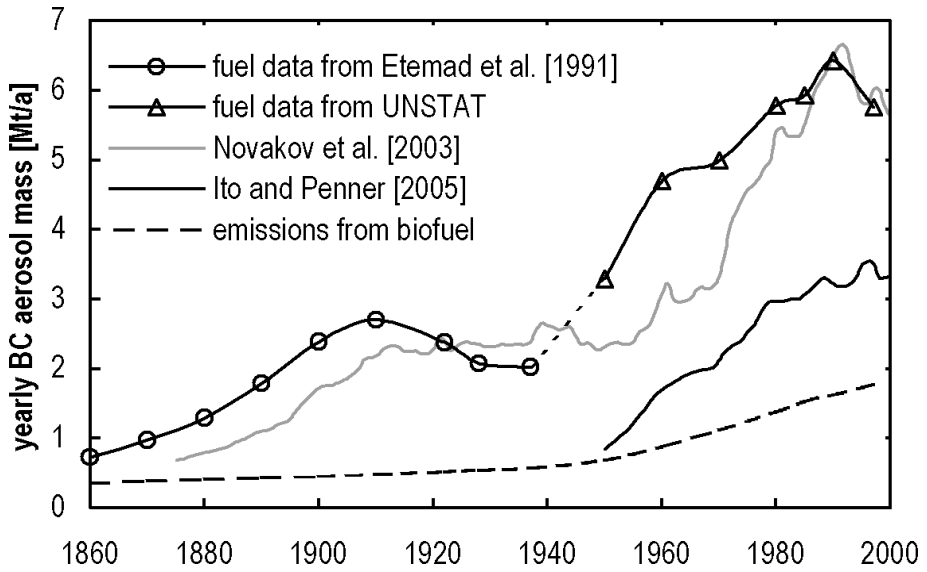

Fig. 8. Total global BC aerosol emissions for the period 1860 to 1997 on the basis of fossil fuel calculated from data of Etemad et al. (1991) and UNSTAT including biofuel. The portion of emissions from Biofuel is also shown. Emissions from fossil fuel given by Novakov et al. (2003) and Ito and Penner (2005) are shown for comparison.

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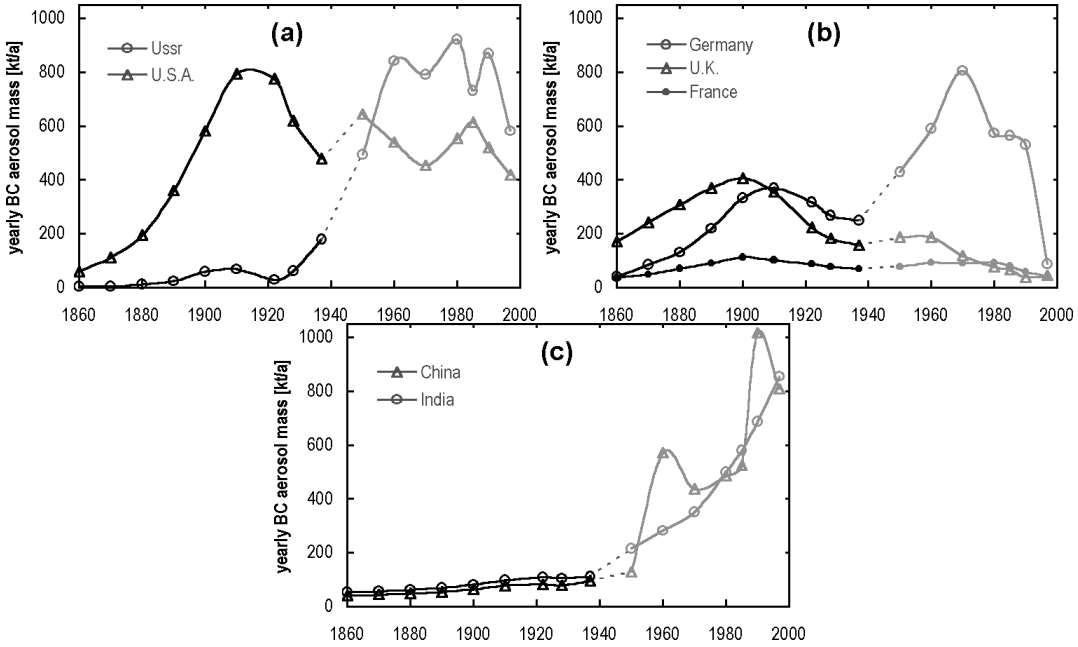

Fig. 9. BC aerosol emissions for the period 1860 to 1997 by country on the basis of fuel production calculated from data of Etemad et al. (1991) (black symbols) and of UNSTAT (grey symbols).

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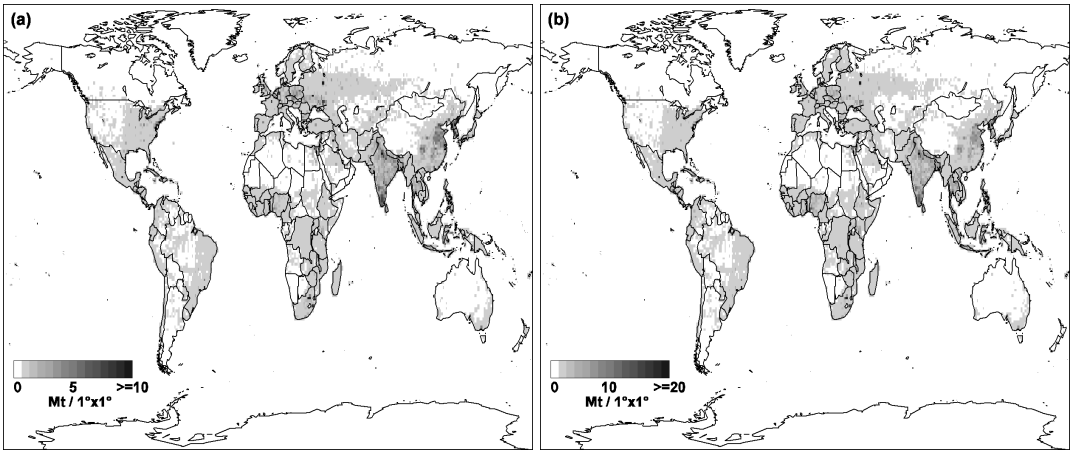

Fig. 10. (a) BC and **(b)** OC emissions for 1997 (fuel consumption from UNSTAT).

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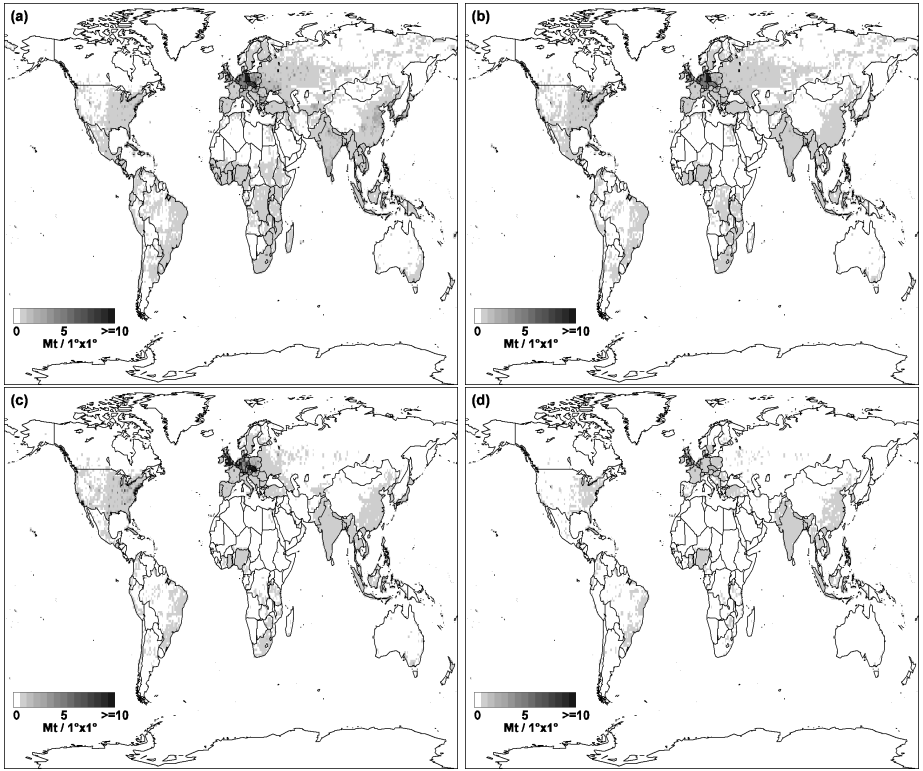

Fig. 11. BC emissions for **(a)** 1970 and **(b)** 1950 (fuel consumption from UNSTAT), and **(c)** 1910 and **(d)** 1860 from Etemad et al. (1991).

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